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*Ernest O. Lawrence*

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RADIOACTIVITY ASSOCIATED WITH  
UNDERGROUND NUCLEAR EXPLOSIONS

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RADIOACTIVITY ASSOCIATED WITH UNDERGROUND  
NUCLEAR EXPLOSIONS

Roger E. Batzel

June 23, 1959

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ABSTRACT

The detonation of a contained or partially contained nuclear explosion is accompanied by the deposition of a large fraction of the energy in the form of high temperature, high pressure regions. The nature of the surrounding medium, the time-temperature history, and the time of cavity collapse or venting determine the extent to which undesirable nuclides such as Sr<sup>90</sup> and Cs<sup>137</sup> will appear outside a fused insoluble matrix and be available to ground water or to the atmosphere. The movement of these undesirable radioactivities relative to the ground water movement can be predicted on the basis of measured K<sub>D</sub>'s (distribution coefficients) for the radioactivities in the medium.

The induced radioactivities are a 20 to 25% contribution to the fission product radioactivity at times the order of one day, a 1% contribution at about 1 week, decreasing to 0.1% at about 45 days, increasing to about 2% because of the Co<sup>60</sup> for a period of 3 to 15 years.

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\* The work was performed under the auspices of the U.S. Atomic Energy Commission.

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Introduction

The detonation of a contained or partially contained nuclear explosion is accompanied by the deposition of a large fraction of the energy in the form of high temperature, high pressure regions. The nature of the surrounding medium, the time-temperature history, and the time of cavity collapse or venting determine the extent to which undesirable nuclides such as  $\text{Sr}^{90}$  and  $\text{Cs}^{137}$  will appear outside a fused insoluble matrix and be available to ground water or to the atmosphere. The movement of these undesirable radioactivities relative to the ground water movement can be predicted on the basis of measured  $K_D$ 's (distribution coefficients) for the radioactivities in the medium.

The induced radioactivities are a 20 to 25% contribution to the fission product radioactivity at times the order of one day, a 1% contribution at about 1 week, decreasing to 0.1% at about 45 days, increasing to about 2% because of the  $\text{Co}^{60}$  for a period of 3 to 15 years.

Explosion Phase

Assuming that a nuclear explosion is fired underground, just subsequent to the prompt energy release from the fission or thermonuclear reactions, the energy of the explosion is deposited in the immediately surrounding cavity at temperatures of the order of a few million degrees.<sup>1</sup> At these temperatures, the chemical behavior of the fission products is unimportant since the matter turns into a hot gas of ionized atoms and electrons.

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<sup>1</sup> G. W. Johnson and C. E. Violet, "Phenomenology of Contained Nuclear Explosions," Lawrence Radiation Laboratory, University of California, Livermore, California, UCRL-5124, Rev. 1, December 1958.

As the shock progresses into the wall of the cavity the first few feet of the surrounding medium is vaporized (about 3 feet for a 2-kiloton explosion) and mixed with the hot gases containing the fission products. As the hot gas cools to a temperature of about 5000°K condensed phases can begin to form, and this condensed phase is then deposited as a part of the molten layer on the wall of the cavity. The majority of the condensation is probably complete in times of the order of milliseconds. Left in the gas phase will be water vapor, noncondensable gases, including rare gases from the fission process and the more volatile fission products and volatile constituents of the medium.

The details of the above phenomena will depend on the time-temperature history, the degree of containment and associated collapse phenomena, and the nature of the medium in which the explosion takes place. If the cavity collapses or vents at a time when the pressure and temperatures are still high, the major fraction of the material in the gas phase will escape from the immediate highly radioactive molten zone.

#### Fission Process

The prompt energy produced in fission including kinetic energy of the fission fragments, prompt gamma rays, and neutrons is 179 Mev.<sup>2</sup> A kiloton is defined as  $1.0 \times 10^{12}$  calories or  $4.185 \times 10^{19}$  ergs and on the basis of 179 Mev per fission, there are  $1.46 \times 10^{23}$  fissions per kiloton. In addition to the prompt energy from fission, there is an average of about 22 Mev of additional energy which results from decay of the fission products. About 50% appears in the form of gamma rays, about 17% as beta decay energy and the other 33% as neutrinos associated with the beta decay process. About 7 Mev of the beta plus gamma decay energy appears in the first 20 minutes after detonation.

The fission of about 55 grams of Pu<sup>239</sup>, U<sup>238</sup>, or U<sup>235</sup> yields an energy equivalent of 1 kiloton. For example a 1-megaton nuclear explosion with a fission-to-fusion ratio of 0.05 has a fission yield of 50 kilotons with its associated 2750 grams of fission products. These 2750 grams of fission products contain about 50 grams of Sr<sup>90</sup> and are the equivalent of the waste fission products from approximately 2 3/4 days' operation of a 1000-megawatt reactor.

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<sup>2</sup> S. R. Gunn, H. G. Hicks, H. B. Levy, and P. C. Stevenson, Phys. Rev. 107, 1642 (1957).

The detonation of a nuclear explosion produces fission products in proportion to the fission yield of the explosion and these fission products are distributed according to the typical fission yield curve shown in Fig. 1. The general characteristics of this fission yield curve are the same for the fission of  $U^{238}$ ,  $U^{235}$ , or  $Pu^{239}$ .

It is necessary to consider the elemental form of the fission products at the time of cavity venting in order to estimate the fraction of the radioactivity which is in the gaseous state. Table I illustrates the contribution of elements as percentages of the total radioactivity at various times after the detonation. These data are taken from the calculations of Bolles and Ballou<sup>3</sup> and apply to products from the thermal fission of  $U^{235}$ .

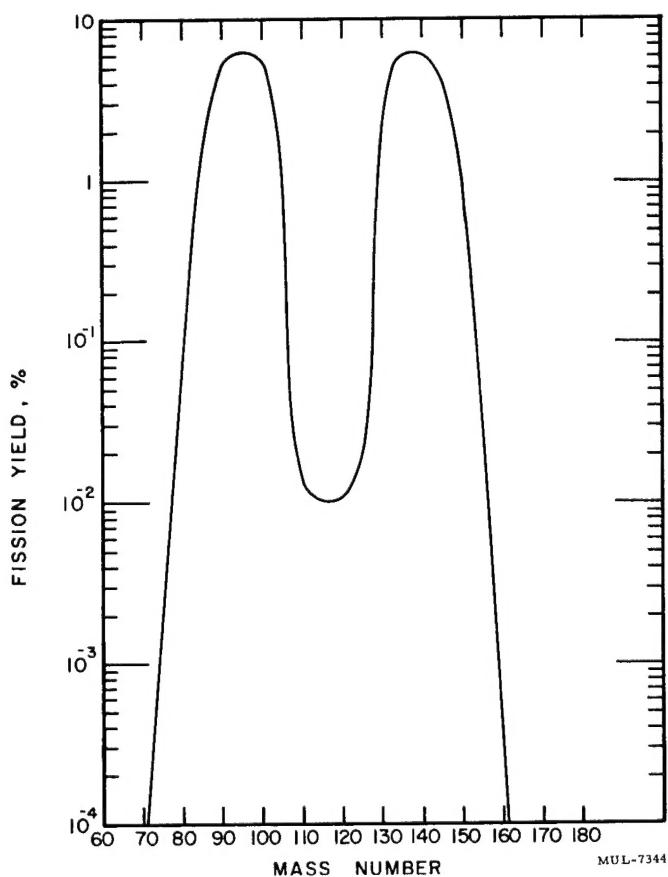


Fig. 1. Yield-mass curve for thermal fission of  $U^{235}$ .

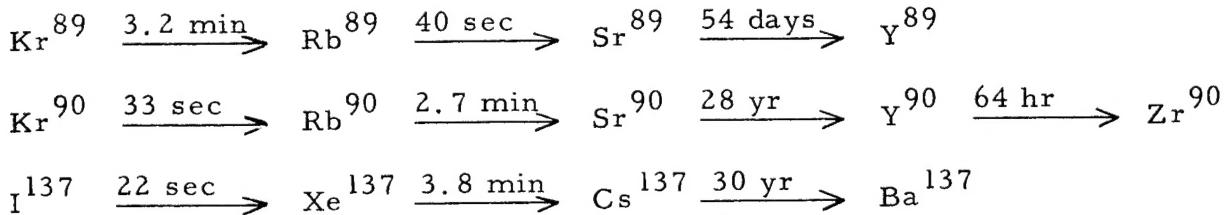
<sup>3</sup> R. C. Bolles and N. E. Ballou, "Calculated Activities and Abundances of  $U^{235}$  Fission Products," USNRDL-456, August 30, 1956.

TABLE I. PERCENTAGE CONTRIBUTION OF ACTIVITY OF ELEMENTS  
TO TOTAL RADIOACTIVITY\*

| Element | 10 sec | 30 sec | 1 min | 5 min | 30 min | 1 hour |
|---------|--------|--------|-------|-------|--------|--------|
| Kr      | 3.8    | 5      | 5.8   | 4.3   | 2.5    | 4.2    |
| Xe      | 2.6    | 3.8    | 4.6   | 9     | 5      | 3.5    |
| Y       | 11     | 9      | 8     | 4.6   | 7      | 5.0    |
| Zr      | 12     | 7      | 3.5   | --    | --     | --     |
| Ba      | 7      | 2.6    | 2.4   | 8     | 9      | 8      |
| Sr      | 6      | 4.0    | 5     | 11    | 4.0    | 3.7    |
| Cs      | 7      | 7      | 7     | 8     | 8      | 8      |
| Rb      | 7      | 8      | 8     | 11    | 7      | 4.8    |
| La      | 9      | 8      | 4.0   | 2.8   | 9      | 10     |
| Nb      | 10     | 12     | 11    | 8     | 5      | 6      |
| Ce      | 4.7    | 3.0    | 1.0   | 2.5   | 5      | 4.4    |
| Br      | 3.3    | 5      | 6     | 2.2   | --     | 1.0    |
| I       | 4.0    | 6      | 7     | 3.5   | 3.5    | 7.0    |
| Mo      | 3.0    | 6      | 6     | 5     | 7      | 3      |
| Te      | 2.7    | 2.5    | 2.0   | 3.5   | 9      | 14     |
| Se      | 1.5    | 1.0    | --    | --    | --     | --     |
| Pr      | 1.7    | 3.7    | 5     | 4.6   | 3.7    | 5      |
| Sn      | 1.1    | --     | --    | --    | --     | --     |
| Tc      | 1.2    | 4.0    | 8     | 5     | 9      | 7      |
| Sb      | 1.0    | 2.3    | 3.7   | 5     | 2.3    | 2.0    |
| Nd      | --     | --     | --    | --    | --     | 1.0    |

\* Radioactivities contributing less than 1% of the total radioactivity are not included.

It is obvious from Table I that a large fraction of the radioactivity exists in the form of noble gases or somewhat volatile elements on time scales which should apply to situations in which cavity venting would occur. The noble gases will not condense until decay to another element has occurred, and the more volatile elements will not condense until the temperature is lowered, or a decay to a more refractory element occurs. The following are typical examples of fission product decay chains:



As an example for the mass-90 chain which is formed in fission, most of the material which finally exists as  $\text{Sr}^{90}$  is produced in the primary fission as  $\text{Kr}^{90}$  (about 80% with smaller amounts formed directly as  $\text{Br}^{90}$  or  $\text{Rb}^{90}$ ). Thus, if the cavity vents in times comparable to the half life of the  $\text{Kr}^{90}$  a major fraction of the final  $\text{Sr}^{90}$  will escape the molten material along with the other gases.

Measurements made on debris from several of the explosions at the Nevada Test Site yield the following typical information:<sup>4</sup>

Strontium-89 — 3-10% in the major part of the fused tuff, enriched in the rubble by several-fold and in the vented debris by nearly an order of magnitude.

Strontium-90 — 20-30% in the fused tuff, vented debris enriched by 5-fold, local fallout less enriched.

Yttrium-91 — 30-50% in the fused tuff, vented debris and rubble enriched several-fold.

Zirconium-95 — 70-100% in the fused tuff, rubble enriched.

Cesium-137 — 20-40% in the fused tuff, vented material enriched by several-fold.

Barium-140 — 30-60% in fused tuff decreasing as yield increases, vented debris enriched several-fold.

Cerium-144 — ~ 100% in the fused tuff.

Neodymium-147 — ~ 100% in the fused tuff, precursors are rare earths.

#### Completely Contained Explosions

Based on the above discussion, even with the formation of an insoluble glassy matrix in which the major fraction of the fission products is incorporated, a large fraction of the  $\text{Sr}^{90}$  and  $\text{Cs}^{137}$  --that fraction which exists as a rare gas at the time of cavity collapse--will not be incorporated but will be distributed

<sup>4</sup> R. H. Goeckermann, University of California, Lawrence Radiation Laboratory, private communication.

with the other gases at the time the cavity vents. Thus appreciable amounts of Sr<sup>90</sup> and other nuclides with rare gas precursors or nuclides of volatile elements would be expected to be deposited at appreciable distances from the highly radioactive zone and to be distributed on the surface of the broken medium as decay products of the rare gas precursors.

#### Partially Contained Explosions

Using the same reasoning which was applied to contained nuclear explosions, depending on the time and extent of venting of a partially contained explosion, the fraction of radioactive debris which does vent to the atmosphere should be rich in the rare gas and more volatile fission products. Sr<sup>90</sup> and Cs<sup>137</sup> should appear enriched with respect to the nuclides which do not have precursors which exist as rare gases at the time the radioactive debris is vented to the atmosphere.

For example, samples of the vented debris from Blanca (20-kiloton) detonation which vented approximately 0.3 to 0.5% of the total fission activity was enriched by about 5-fold in Sr<sup>90</sup> indicating that about 1.5 to 2.5% of the total Sr<sup>90</sup> formed in the explosion was actually released to the atmosphere. As the rare gases decay to atoms such as Rb<sup>90</sup> and subsequently to Sr<sup>90</sup>, these atoms would be expected to deposit rapidly upon the surface of the large amount of particulate matter which is blown out with the vented gas and to be scavenged from the atmosphere as the particulate matter falls out.

#### Ground Water Contamination

From the above discussion, it follows that the formation of an insoluble glassy material during the explosion does not guarantee the trapping of all the undesirable radioactivities. In fact, large fractions of two of the more undesirable nuclides from a biological point of view, Sr<sup>90</sup> and Cs<sup>137</sup>, will appear outside the fused material and in the broken material and regardless of arguments of the existence of fused, glassy, insoluble material in which the majority of the other fission products are contained, these nuclides will be accessible to ground water.

Higgins<sup>5</sup> in his paper on ground water discussed the fact that ions such as Sr<sup>90</sup> and Cs<sup>137</sup> are selectively absorbed from ground water by most naturally

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<sup>5</sup> G. H. Higgins, "Evaluation of the Ground Water Contamination Hazard from Underground Nuclear Explosions," Lawrence Radiation Laboratory, University of California, Livermore, California, UCRL-5538, April 8, 1959.

occurring minerals and that these ions distribute themselves between water and minerals according to the following law

$$K_D = \frac{A_S \times M_w}{A_w \times M_S}$$

where  $K_D$  is the distribution coefficient,  $A_S$  and  $A_w$  are the activities in the mineral and water respectively, and  $M_w$  and  $M_S$  are the masses of water and mineral respectively in equilibrium. Given  $K_D$  of a nuclide, it is possible to calculate the average flow rate of the activity as a function of the ground water flow rate.

$$F_A = F_w \frac{1}{1 + K_D \rho}$$

where  $\rho$  is the ratio of the mass of mineral per unit volume to the mass of water per unit volume.  $\rho = 10$  represents the typical situation in most situations in which ground water occurs.  $K_D$ 's, distribution coefficients, for natural occurring minerals range from minima of about 100 (limestone) for  $Sr^{90}$  to 100,000 for  $Ce^{144}$ . With a typical flow rate for ground water of 3 feet per day for example, assuming  $K_D = 300$  and  $\rho = 1$ , the average flow rate of  $Sr^{90}$  in the medium would be

$$F_A = 3 \text{ ft/day} \times \frac{1}{1 + 300} = 0.01 \text{ ft/day}$$

so that in a time equal to the half life of  $Sr^{90}$  (28 years) the  $Sr^{90}$  activity would have moved about 100 feet. See the Appendix for an example of the effect of the distribution coefficient on the concentration of  $Sr^{90}$  in ground water.

On the basis of the above considerations, it is possible by having a detailed knowledge of the hydrology of an area and by making laboratory measurements on minerals which will be in contact with the ground water of the  $K_D$ 's (distribution coefficients) for the biologically undesirable nuclides to predict the fate of these radioactivities in the ground water. In general, because of the ion exchange characteristics of naturally occurring media, ground water contamination does not appear to present any serious hazard, nor does the ground water contamination problem appear to place any serious limitations on the use of nuclear explosives.

#### Induced Radioactivities

Based on the assumption that  $\sim 1 \times 10^{23}$  neutrons per kiloton of yield are emitted from a nuclear explosion and that these  $1 \times 10^{23}$  neutrons are captured

in the surrounding medium, it is possible to estimate the levels of radioactivity induced in a typical medium surrounding a nuclear explosion. The major fraction of the induced radioactivity would be expected to occur in the first 2 or 3 feet of medium and to be included in the initially vaporized material.

Table II gives the typical medium composition chosen and it is the same as that given in Brian Mason, Principles of Geochemistry, John Wiley and Sons (1952). For comparison, the results of an analysis of Nevada sand is shown. Oxygen is not included since the oxygen absorbs substantially less than 1% of the neutrons. The composition in the case of the typical medium includes 20% by weight water.

TABLE II. MEDIUM COMPOSITION

| <u>Typical Medium</u> |                   | <u>Nevada Sand</u> |                   |
|-----------------------|-------------------|--------------------|-------------------|
| Element               | Abundance by Wt % | Element            | Abundance by Wt % |
| Si                    | 50                | Ca                 | ~ 30              |
| Al                    | 14.5              | Si                 | ~ 25              |
| Fe                    | 9                 | Al                 | ~ 20              |
| Ca                    | 6.4               | Fe                 | ~ 10              |
| H                     | 5.1               | Na                 | ~ 8               |
| Na                    | 4.9               | Mg                 | ~ 2               |
| K                     | 4.7               | K                  | ~ 3               |
| Mg                    | 3.7               | Ti                 | 0.1 - 1%          |
| Ti                    | 0.6               | B, Mn, Cu          | 0.01 - 0.1%       |
| P                     | 0.18              |                    |                   |
| Mn                    | 0.18              |                    |                   |
| Co                    | 0.0042            |                    |                   |

The neutrons captured in the typical medium were distributed according to the atom percent of the element present and the thermal neutron capture cross sections for the element.<sup>6</sup> With 20% by weight water, most of the neutrons are thermallized before capture occurs, and about 60% of the neutrons

<sup>6</sup> D. J. Hughes and R. B. Schwartz, "Neutron Cross Sections," Brookhaven National Laboratory, Upton, New York, BNL-325, July 1, 1958.

are captured by the hydrogen of the water in this typical medium. The important radioactive nuclides produced and the percent of the neutron capture leading to these nuclides is shown in Table III.

TABLE III. RADIOACTIVE NUCLIDES PRODUCED

| Element                         | Nuclide Produced | Half Life <sup>7</sup> | Gamma Energy <sup>7</sup> | Percent of Neutron Captures leading to Nuclide |
|---------------------------------|------------------|------------------------|---------------------------|--|
| Na                              | Na <sup>24</sup> | 15 hours               | 4.0 Mev                   | 3.7%   |
| Al                              | Al <sup>28</sup> | 2.3 min                | 1.8 Mev                   | 2.6%   |
| Mn                              | Mn <sup>56</sup> | 2.6 hours              | 1.8 Mev                   | 1.3%   |
| Fe<br>(0.33% Fe <sup>58</sup> ) | Fe <sup>59</sup> | 45 days                | 1.3 Mev                   | 0.015%   |
| Co                              | Co <sup>60</sup> | 5.2 years              | 2.5 Mev                   | 0.09%  |

In Table IV are listed the number of curies per kiloton of these nuclides estimated to be produced.

TABLE IV. CURIES PER KILOTON OF NUCLIDES PRODUCED

| Nuclide          | Curies            | (Curies) × (Gamma Energy) |
|------------------|-------------------|---------------------------|
| Na <sup>24</sup> | $1.3 \times 10^6$ | $5.2 \times 10^6$         |
| Al <sup>28</sup> | $3.5 \times 10^8$ | $6.3 \times 10^8$         |
| Mn <sup>56</sup> | $2.6 \times 10^6$ | $4.7 \times 10^6$         |
| Fe <sup>59</sup> | $\sim 10^2$       | $\sim 1.3 \times 10^2$    |
| Co <sup>60</sup> | $\sim 10$         | $\sim 25$                 |

The number of (curies) × (gamma ray energy) is a measure of the relative biological effectiveness of the gamma rays. Figure 2 is a comparison of these radioactivities as a function of time with the fission product radioactivities from 1 kiloton of fission yield. The induced radioactivities are a 20 to 25% contribution to the fission product radioactivity at times the order of one day, a 1% contribution at about 1 week, decreasing to 0.1% at about 45 days, increasing to about 2% because of the Co<sup>60</sup> for a period of 3 to 15 years.

<sup>7</sup> D. Strominger, J. M. Hollander, and G. T. Seaborg, Rev. Modern Phys. 30, 585 (1958).

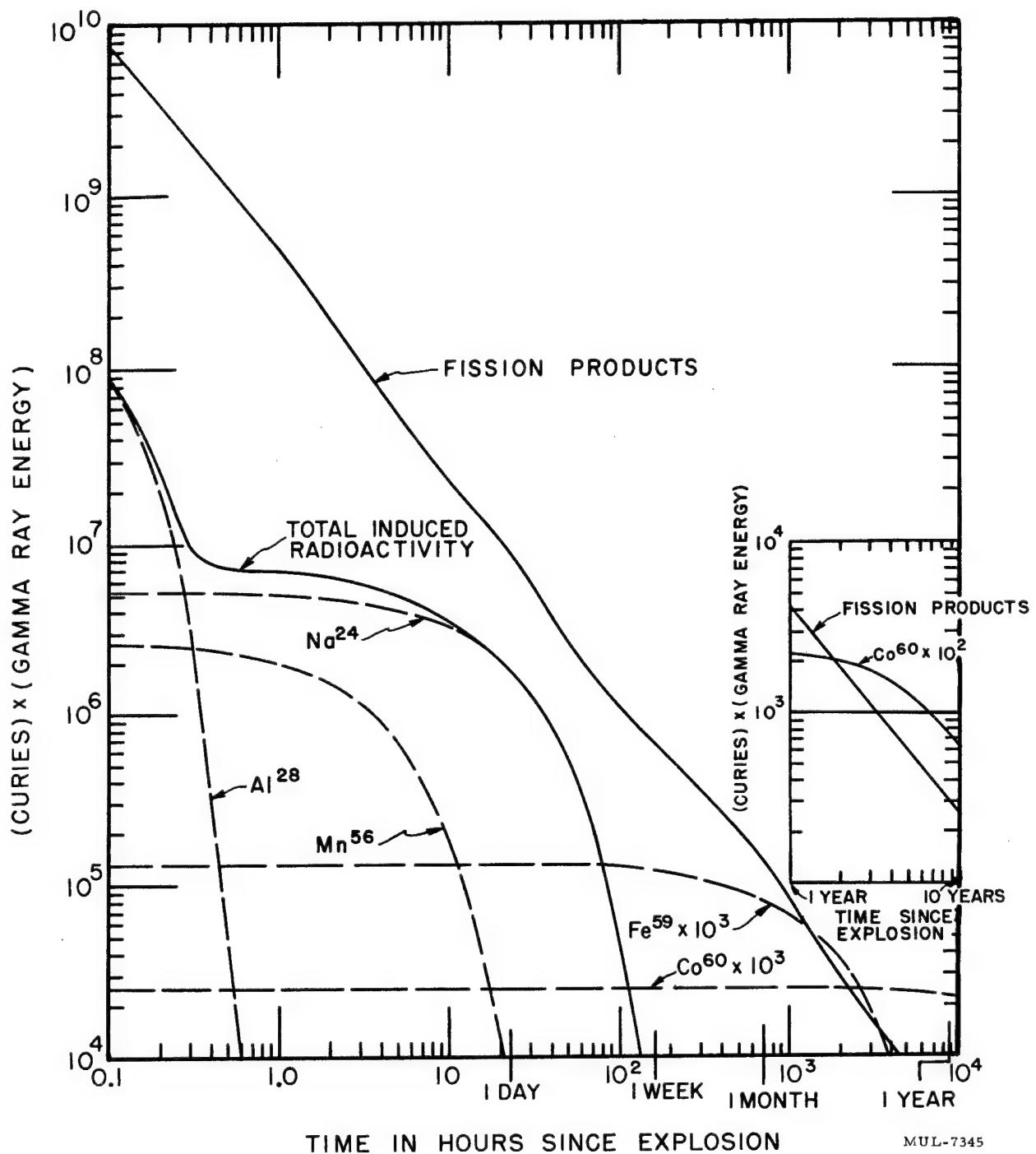


Fig. 2. Comparison of induced radioactivity and fission product radioactivity from a one kiloton nuclear explosion.

Appendix

Example of Distribution of Sr<sup>90</sup> from a Nuclear Explosion

If it is assumed that all of the Sr<sup>90</sup> escapes from the cavity and is distributed in an amount of broken medium equivalent to the fused material as observed in Nevada Tuff (~ 500 tons of fused material per kiloton), the concentration of Sr<sup>90</sup> in the broken medium is, on the basis of an explosion with 100% fission, 1 gram or ~ 150 curies of Sr<sup>90</sup> per kiloton of fission, and the 1 gram distributed in 500 tons of medium,

$$\frac{150 \text{ curies}}{500 \text{ tons} \times 10^6 \text{ g/ton}} = \sim 3 \times 10^{-7} \text{ curies/g}$$

The actual situation is probably better represented by the distribution of the 150 curies of Sr<sup>90</sup> in some 5000 to 50,000 tons of broken medium.

If a typical K<sub>D</sub> of 300 is used, the concentration of Sr<sup>90</sup> in ground water in contact with the medium is

$$\frac{3 \times 10^{-7} \text{ curies/g}}{300} = 1 \times 10^{-9} \text{ curies/g}$$

of water. After the 1 gram or milliliter of ground water has equilibrated with an uncontaminated gram of the medium the concentration of Sr<sup>90</sup> in the ground water is  $\frac{1.0 \times 10^{-9} \text{ curies/gram}}{300}$  or ~ 3.3 micromicrocuries/gram of water or about 10 times the tolerance level. It is clear that equilibration of the 1 gram of the water with another gram of medium will leave the concentration of Sr<sup>90</sup> below the tolerance level.

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LIST OF PREVIOUS PLOWSHARE REPORTS

|                     | Title   |
|---------------------|---|
| UCRL-4659           | Deep Underground Test Shots   |
| UCRL-5026           | Non-Military Uses of Nuclear Explosions   |
| UCRL-5124<br>Rev. I | Phenomenology of Contained Nuclear Explosions   |
| UCRL-5253           | Industrial Uses of Nuclear Explosives   |
| UCRL-5257<br>Rev.   | Peaceful Uses of Fusion   |
| UCRL-5281           | Temperatures and Pressures Associated with the Cavity<br>Produced by the Rainier Event.                 |
| UCRL-5457           | Large Scale Excavation with Nuclear Explosives  |
| UCRL-5458           | Mineral Resource Development by the Use of Nuclear<br>Explosives  |
| UCRL-5538           | Evaluation of the Ground Water Contamination Hazard from<br>Underground Nuclear Explosions              |
| UCRL-5542           | Properties of the Environment of Underground Nuclear<br>Detonations at Nevada Test Site. Rainier Event. |

## DISTRIBUTION OF VENTED DEBRIS

Roger E. Batzel

October 30, 1959

The fraction of total radioactivity which vents to the atmosphere is a function of scaled depth of burial. The actual functional relationship is uncertain, but data are available for several scaled depths:

| Event      | Yield (KT)                                       | <u>Vented Radioactivity</u> |  | Measured Radioactivity Deposited on Surface % |
|------------|--|-----------------------------|--|---|
|            |  | Scaled Depth (feet)         |  |   |
| Jangle-U   | 1.2 $\pm$ 0.1                                    | 16                          |  | > 80  |
| Teapot-Ess | 1.2 $\pm$ 0.1                                    | 63                          |  | 90  |
| Neptune    | 0.090 $\pm$ 0.020                                | 220                         |  | 1 - 2   |
| Blanca     | 19.0 $\pm$ 1.5                                   | 310                         |  | < 0.5   |
| Logan      | 5.0 $\begin{matrix} + 0.2 \\ - 0.4 \end{matrix}$ | 485                         |  | 0   |

The region of uncertainty in the fraction of radioactivity vented to the atmosphere is at scaled depths between 63 feet and 220 feet.

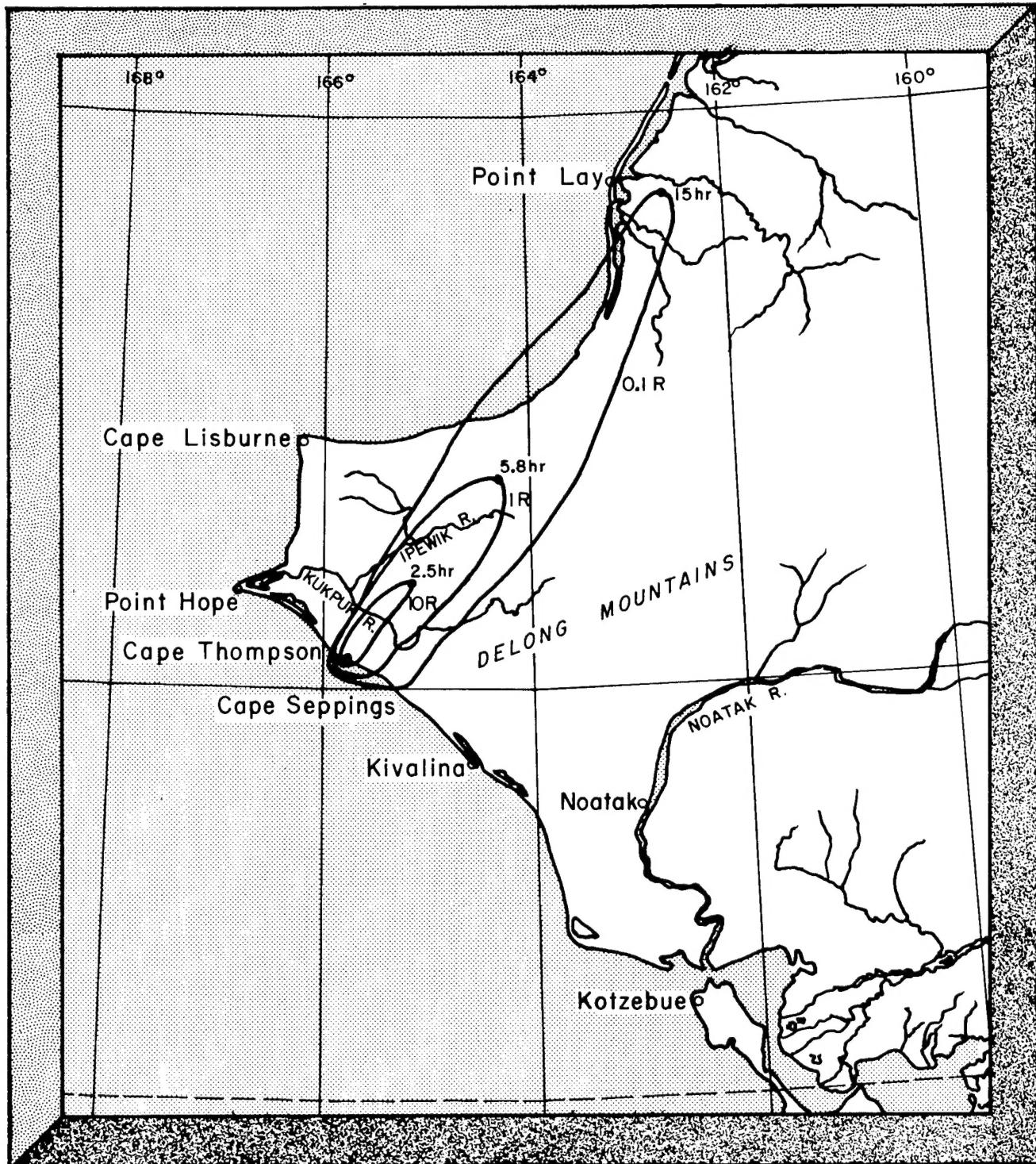
The major fraction of the vented radioactivity is deposited locally from partially contained nuclear explosions (i.e. excavation applications). The degree of localization is also expected to be a function of scaled depth of burial for at least two reasons. First, the mass of material involved in the explosion which contacts the vented radioactivity increases as the scaled depth of burial is increased and the average particle size to which the radioactivity attaches itself should increase. Thus the fall rate of the particles containing the radioactivity should be greater with greater scaled depth. Second, the fraction of total thermal energy which escapes with the radioactivity

should decrease as the depth of burial increases. This means that the average altitude to which the post explosion debris cloud rises for a given total yield should decrease as the scaled depth of burial increases. The combination of these two effects, higher particle fall rates and lower debris cloud heights, work together to localize the deposition of the vented radioactivity as the scaled depth of burial is increased.

In addition to the above effects which tend to localize the radioactivity, there is another effect which should lower the amount of radioactivity which escapes to the atmosphere as the total yield is increased and the scaled depth of burial remains constant. At a constant scaled depth, the depth of burial increases as  $W^{1/3}$  ( $W$  is the yield). Thus the path length for the radioactivity to reach to the surface increases with  $\sim W^{1/3}$ , and the increased amount of matrix material which the venting radioactivity contacts should tend to remove a proportionally larger fraction of the radioactivity.

Figure 1 is a typical fallout pattern which was calculated by A. V. Shelton as representative for the proposed Chariot experiment. The assumptions are that fraction of total radioactivity escaping is 5%, that the debris cloud height is 30,000 feet, and that the fall rate of the particles is about 4 times the fall rate for particles from an explosion on the surface of the ground. A wind pattern which would represent an acceptable condition for the explosion was chosen as a basis for the calculation.

The  $Sr^{90}$  and  $Cs^{137}$  content of the fission product debris is enriched by a factor of about 5-fold over the usual fission yield distribution. However because of the low debris cloud heights and the large mass of matrix material, essentially none of the  $Sr^{90}$  and  $Cs^{137}$  should enter into the world-wide fallout system.



TYPICAL FALLOUT PATTERN, CHARIOT

Infinite Dose, Roentgens

5% vented debris

Winds of 4 April, 1959

Cloud Height 30,000'

Cloud radii 4 and 2 n.mi

MUL-7971